ARKIV FÖR FYSIK Band 1 nr 24

Communicated 9 March 1949 by MANNE SIEGBAHN

The disintegration of Sb^{125} and Te^{125*}

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With 6 figures in the text

When Sn is irradiated with slow neutrons a number of active Sn isotopes with different half lives are created. Listed in order of increasing massnumber, the more important isotopes are[1]: Sn^{113} (105^d), Sn^{121} (28^h), Sn^{123} (10^d) and Sn^{125} (10^m). All are β-emitters except Sn^{113}, which decays by K capture. They decay to the corresponding Sb isotopes. These latter are all stable except Sb^{125}, which is known[1] to have a half life of about 2.7 years and to decay by β and γ-emission to Te^{125}. Furthermore this latter isotope possesses an isomeric state ($\tau \sim 60$ days) at ~ 120 KeV, which can also be obtained by K capture of I^{125}.

Until quite recently the only information on the mode of disintegration of Sb^{125} and Te^{125*} were based on absorption measurements. Thus, in the Plutonium Project literature[1] C. STANLEY and L. GLENDENIN have reported the existence of two β-components of the energies 0.3 MeV (65 %) and 0.7 (35 %) in the radiation from Sb^{125} (abs. in Al). The radiation also contains a γ-component of 0.6 MeV (abs. in Pb). The absorption curve for the internal conversion electrons from the Te^{125*} metastable state was studied by FRIEDLANDER, GOLDHABER and SCHARFF-GOLDHABER[2]. Shortly before the results of the present investigation were published two more papers appeared covering the same subject using spectrometer technique. One of these was by KERN, MITCHELL and ZAFFARANO[3] (Beta- and Gamma-Rays of Antimony 125) and the other by HILL, SCHARFF-GOLDHABER and FRIEDLANDER[4], (Internal Conversion Electrons from Metastable Te^{125}). Many of their results agree quite well with those which will be presented in this paper, but additional data have been obtained in the present investigation, which enable a more complete discussion of the complicated termscheme for the Sb-Te*-disintegration.

Chemical preparation

The active Sb and Te* (in equilibrium with Sb) was obtained from the Oak Ridge Pile. The irradiated sample consisted of 6 g of metallic Sn. The Sb and Te activities were small and had to be separated from the total material, which mainly contained the strong Sn^{113} activity. The first part of the investigation was concerned with finding a method for extracting the Sb and Te* fractions quantitatively and free from other activities. In order to get high specific activities it was necessary to do this by using only minute amounts of carriers. It was found that the following procedure was convenient.

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[1] For earlier works see: G. SEABORG and I. PERLMAN, Table of Isotopes, Review of Modern Physics, *20*, 1948.

[2] G. FRIEDLANDER, M. GOLDHABER and G. SCHARFF-GOLDHABER, Phys. Rev. *74*, 981, 1948.

[3] B. KERN, A. MITCHELL and D. ZAFFARANO, Communication at the American, Phys. Soc. January 1949 Meeting.

[4] R. HILL, G. SCHARFF-GOLDHABER and G. FRIEDLANDER, Phys. Rev. *75*, 324, 1949.

Principle of separation

The irradiated tin was dissolved in hydrochloric acid and a carrier solution of indium and tellurium added. Tellurium was precipitated as metal by reduction with sulphur dioxide and hydrazine hydrochloride. The main part of the active antimony was found in the filtrate to which a small amount of antimony was added as a carrier. The antimony was separated from the tin with hydrogen sulphide in 2N hydrochloric acid followed by a second purification using hydrogen sulphide in the presence of oxalic acid.

Experimental

About 3.5 g of the irradiated tin was dissolved in 4–5 ml of hot HCl with a few crystals of potassium chlorate. To this solution a carrier solution of indium (1 mg) and tellurium (3 mg) in 1 ml HCl was added. HNO_3, used to dissolve the carriers, had previously been removed by repeated evaporation to dryness with HCl.

When the solution had been diluted to 25 ml, the tellurium was precipitated from the boiling hot solution by means of 7 ml of a freshly prepared saturated solution of SO_2 followed by 5 ml of a 15 % aqueous solution of hydrazine hydrochloride and finally an additional 12 ml of the saturated SO_2 solution. After the solution had been boiled for a few minutes, the precipitate settled was filtered off and thoroughly washed with hot water.

The filtrate was evaporated almost to saturation and the tellurium reprecipitated in about 3 N HCl. Before the filtration the solution was kept hot on a water bath for some hours. When this small amount of tellurium had been removed no further black precipitation was visible at the evaporation of the filtrate.

The tellurium precipitates were dissolved in 4 ml HCl with a few crystals of potassium chlorate. A carrier solution of antimony (1.5 mg) and tin (2.5 mg) in 0.5 ml HCl plus 5 ml H_2O was added and the tellurium precipitated with hydrazine hydrochloride in presence of tartaric acid.[1]

The filtrate was evaporated and tested for tellurium, but no precipitation was now visible.

The tellurium was dissolved and reprecipitated twice in the same manner before the β-spectrograph measurements were started. Each time small quantities of inactive antimony and tin in HCl-solution were added.

The filtrate, after the precipitation in the presence of tartaric acid, contained a small amount of antimony and tin. The antimony was separated from the tin by precipitation with H_2S in a solution, which contained oxalic acid.[2] The sulphide precipitation from this separation carried only a small fraction of the Sb-activity, but was used as carrier for the filtrate containing the irradiated tin and the main part of the antimony activity.

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[1] A. Gutbier, Z. anorg. Chem. *32*, 264, 1902.

[2] W. W. Scott, Stand. Meth. Chem. Anal., New York 1939, I, 68.

The last mentioned filtrate, which contained large quantities of tin, was first purified from traces of tellurium, and then cautiously evaporated almost to dryness. (It should be noticed, that when HCl-solutions containing antimony are evaporated, there is always a great risk of losing antimony.) 25 ml H_2O and 6 ml HCl were added and part of the antimony was separated from the tin by adding H_2S to the hot solution. The precipitate was filtered off and washed with hot water. In order to get both a quantitative yield of the small amount of antimony and a good separation from most of the relatively large amount of tin, it was found that a good method was a fractionated precipitation, where the filtrate from the first precipitation was diluted with about 5 ml of H_2O and reprecipitated with H_2S in the same way as before. The new filtrate was diluted with 5 ml H_2O and precipitated etc.[1] By measuring the activities it was decided in which of the precipitations the antimony was present. These precipitations were dissolved and finally the antimony was separated from the remaining tin by two precipitations with hydrogen sulphide in the presence of oxalic acid and inactive tin.

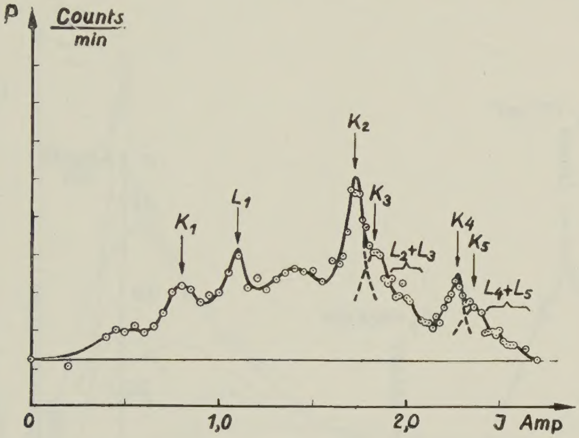

Fig. 1. Photo electron spectrum from Sb^{125} with a 0.02 mm Pb converter.

The sulphide precipitate was dissolved in a little aqua regia evaporated to dryness at 60° C and redissolved in a drop of diluted HCl.

The γ-radiation of Sb^{125}

The γ-radiation was studied in a lens spectrometer by means of the secondary photo electron spectrum obtained from a 0.02 mm lead converter. The total activity was only 4.5 μC. Ra γ-equivalence (measured with a gold cathode γ-tube) and the resulting poor statistics made it necessary to run the spectrum several times. One of these photo electron spectra is shown in Fig. 1. The resolution of the spectrometer was set to be 4.5 % in order to get high transmission. One can distinguish 5 different γ-lines. The first two photo lines correspond to the K and L photoline of a γ-ray of 175 KeV. They are of almost the same intensity, which can be understood because of the much shorter range of the 87 KeV K-photo electrons compared to the 159 KeV L-photo electrons in the lead converter. The next photo line is double and so is the last one. This was evident from all independent runs. The energy of the first of these γ-lines is 425 KeV and the second ~ 465 KeV. The intensity ratio between these two γ-lines is roughly 3 : 1. The forth γ-line has an energy of 601 KeV and the fifth ~ 637 (intensity ratio ~ 2 : 1). The L-photo electrons of these four lines can be observed but the line character is smeared out. The exact values of the 465 and 637 KeV γ-lines are less certain but their existence can not be doubted since the half widths of the remaining photo lines otherwise would be too great.

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[1] Comp. A. A. Noyes and W. C. Bray, J. Am. Chem. Soc. 29, 203–4, 1907.

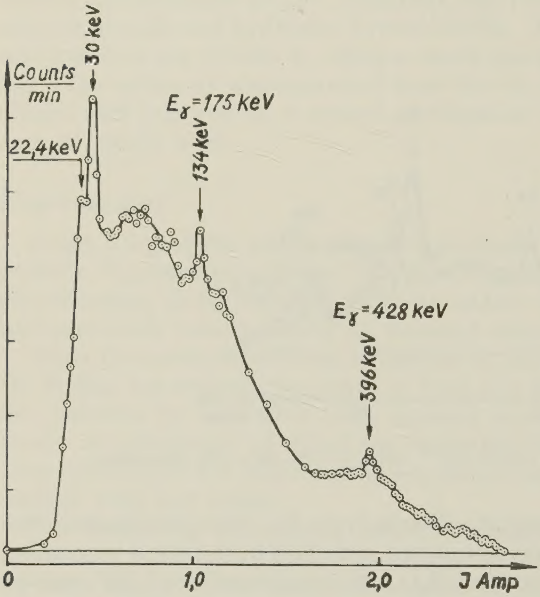

Fig. 2. Sb^{125} β-spectrum (undivided form).

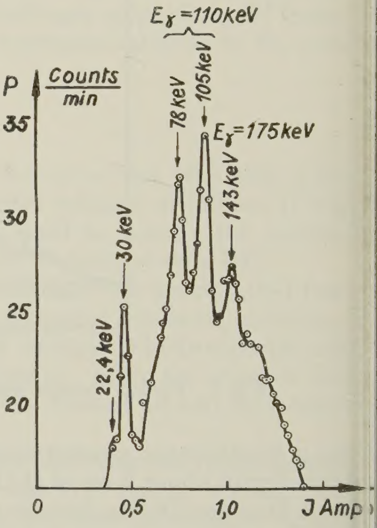

Fig. 3. Part of the Sb^{125} β-spectrum with an aged sample.

The energies of the γ-lines obtained from the photo spectrum agree very satisfactory with those obtained by KERN et al. (loc. cit.). They report the following γ-energies: 172, 432, 478, 614, and 638 KeV compared to our: 175, 425, 465, 601 and 637 KeV.

β-spectrum

The β-spectrum was investigated in the same spectrometer. Fig. 2 shows one of the several runs. For convenience the spectrum is given in undivided form, e. g. to get the true intensity from the curve one has to divide it by the corresponding I-value. In addition to the one conversion line corresponding to the γ-line at 175 KeV and the one at 428 KeV (from the photo spectrum 425 KeV) two very soft lines at 22 KeV and 30 KeV can be seen. Low intensity β-lines between the 30 KeV and the β-line corresponding to $E_\gamma = 175$ KeV cannot be excluded, but if they exist they are of much smaller intensities than the others. If the sample is aged for some time and then investigated again two rather strong lines appear in this region at energies of 78 KeV and 105 KeV. They belong to the isomeric state of Te^{125*}, which continuously grows from Sb^{125}. If the Te is precipitated from the sample the two lines disappear again. Fig. 3 shows this part of the β-spectrum taken with a sample, aged for about 1 month. It will later be seen that the energies of the two extra β-lines agree with those of Te^{125*}.

After division the continuous spectrum in fig. 2 was treated by Fermi analysis in order to resolve it into different β-components. Convenient diagrams for the evaluation of the appropriate functions used in these analyses have been given by Bleuler and Zünti.[1] The Kurie plots will not be reproduced here; instead the spectrum after division and separation into components is shown in Fig. 4. The spectrum can be resolved into three partial spectra with the upper limits 616 KeV, 299 KeV and 128 KeV. The intensities of these components are 18 %, 49 % and 33 %. Kern et al. (loc. cit.) have found two components with $E_{\beta_{\max}} = 659$ KeV and 281 KeV but suggest that one or more low energy components may also be present. Except for the two soft internal conversion lines, which were not observed, the 174 KeV and 431 KeV were also reported by Kern et. al. in agreement with the results obtained here. Kern found, in addition, two β-lines, corresponding to the γ-energies 110 KeV and 125 KeV. It is possible that these latter lines do not belong to the primary Sb disintegration but are identical with the K and L lines of Te* as shown in Fig. 3.

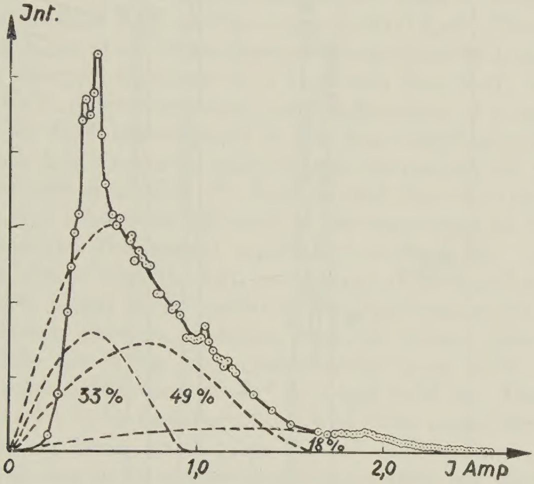

Fig. 4. Sb^{125} β-spectrum resolved into three different β-components.

Internal conversion lines of Te^{125*}

The metastable Te fraction was chemically isolated from Sb according to the above described method. Though the total activity was rather poor the specific activity was very high and made it possible to study the electron distribution down to very low energies. This was done in two different spectrometers, one provided

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[1] E. Bleuler and W. Zünti, Helvetica Phys. Acta XIX, 375, 1946.

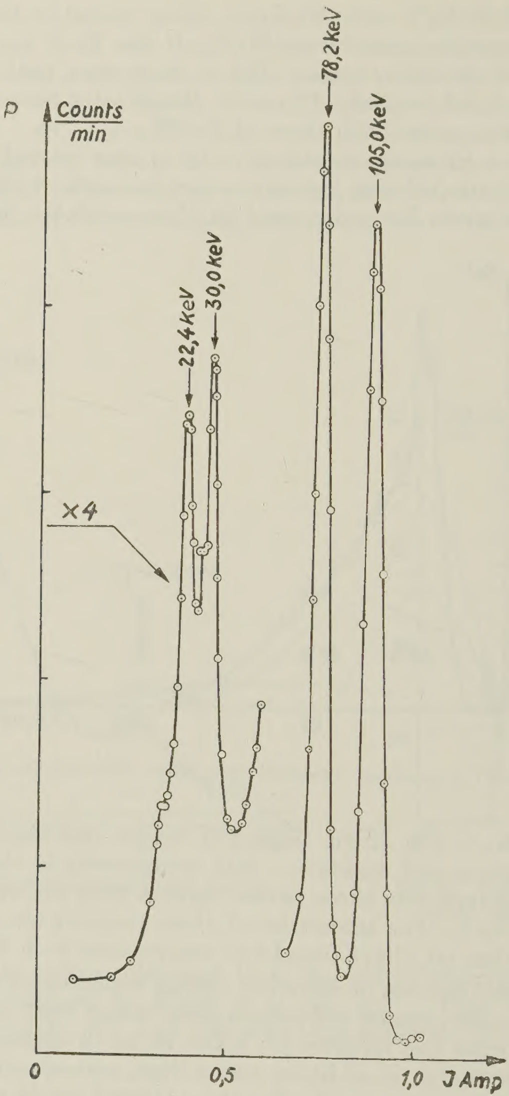

Fig. 5. Internal conversion electron spectrum of Te^{125*}. The two left lines are magnified 4 times.

with a nylon window with a cut off at about 10 KeV and the other with a grid-supported zaponlac window with a cut off at about 2 KeV.

Fig. 5 shows the internal conversion spectrum of Te^{125*}. Two strong lines at 78.2 KeV and 105.0 KeV occur, corresponding to a γ-ray energy of 110 KeV. These are the lines responsible for the transition from the metastable state to a lower level in Te^{125}. A high spin change is connected with this transition. This is evident from the small value of the K to L conversion ratio. In order to get high transmission the resolution used here was not sufficient to resolve the L and M lines and it is therefore only possible to give the intensity ratio K: (L+M), which is 1.10. KERN et al. (loc. cit.) reports K : L = 1.2 and HILL et al. (loc. cit.) K : L : M = 1.5 : 1 : 0.3. If the former measurement includes also the M line as in our case the three intensity measurements agree quite well. HILL et al. have pointed out that this intensity ratio would mean a $\triangle l = 5$ and that the transition would consist of 95 % 2^4 pole magnetic and 5 % 2^5 pole electric radiation. The spin change is 4. This calculation is based on a recent theory by DRELL.

According to Fig. 5 two other internal conversion lines of rather small intensity have been found, one at 22.4 KeV and the other at 30.0 KeV. The last of these lines was also observed by HILL et al. If we interpret these lines as L conversion lines the corresponding γ-ray energies would be 27.4 KeV and 35.0 KeV, if they are K lines 54.2 KeV and 66.8 KeV. There are some small indications of a possible third β-line of low intensity at 19 KeV investigated in the zaponlacwindow spectrograph but the intensity was too low to enable any definite statement on this point and its existence is therefore very doubtful. To be sure that the other two low energy lines belong to Te* their half lives were followed in the spectrograph for more than two months and both decayed the correct amount ($\tau = 60$ days).

It is important to notice that the two low energy β-lines and also the two other β-lines (K and L lines) found in Te* occur in the spectrum of Sb as well. A significant difference between them is, however, that the former lines are present in a fresh Sb sample, while the latter lines continuously grow while the Sb sample is aging i.e. when the metastable state of Te* is being built up. Thus the low energy levels are in common for both disintegrations and they must therefore almost certainly be placed *after* the level, which gives the 110 KeV radiation of Te*.

A possibility, which has to be considered here, is that the 22.4 KeV line might very well be an Auger electron line. The K radiation of Te has an energy of 26.9 KeV and is therefore definitely too small to explain the line at 30 KeV as being an Auger line. However if we subtract the L binding energy of the next element of higher Z-value, i.e. iodine, which is 5.2 KeV, we obtain 21.7 KeV, which should correspond to the energy of one of the possible Auger transitions. This energy comes very close to the observed 22.4 KeV. Furthermore one certainly has to expect Auger electron lines in the spectrum due to the very high conversion rate of the 110 KeV transition (probably near 100 %). We may therefore conclude that a reasonable explanation of the 22.4 KeV line is that it is due to an Auger transition of the type K-L-L^1. A small hump in the curve between the two soft lines could possibly correspond to a K-L-M^1 transition, the energy of which should be 25.8 KeV compared to the observed value 26.8 KeV.

Another circumstance which seems to favour this explanation is that the 22.4 KeV line is smaller compared to the 30 KeV in the case of Sb than in Te*. This would be expected since the 175 KeV radiation (and also the higher energy β-lines) in the Sb spectrum is converted much less than the 110 KeV line in Te*. It should be stressed, however, that the intensity ratio between the two lines in the Sb spectrum is rather uncertain, since the 22.4 KeV line at least is situated on the low energy side of a continuous spectrum.

The disintegration scheme

We are now in a position to discuss the total disintegration scheme for Sb and Te*. Altogether there are 7 different γ-lines, obtained from the photo electron spectrum and internal conversion spectrum. The continuous β-spectrum has been resolved into three different components. Furthermore one electron line in the β-spectrum has been interpreted as an Auger electron line. A disintegration scheme which includes all these transitions has been constructed in Fig. 6.

This scheme is based on the following arguments. The soft γ-line of Te*, which is emitted also from Sb has to be in cascade with the 110 KeV transition and leads then to the ground state in Te, if no other γ-rays are supposed to exist in the Te* disintegration. The difference between the γ-rays at 465 KeV and 425 KeV is 40 KeV and between 637 KeV and 601 KeV is 36 KeV. These differences are equal, within the experimental limits to the previously discussed soft γ-ray, if it is assumed that the internal conversion line originates from the L shell. In that case the γ-energy was 35 KeV. The energy of the K internal conversion line is too small to be observed, i.e. 3 KeV. We may therefore conclude that the γ-rays of 465 KeV and 637 KeV lead to the groundstate in Te, whereas the γ-rays of 425 KeV and 601 KeV to the 35 KeV level. Furthermore the sum of the last γ-ray (175 KeV) and 465 KeV is 640 KeV, which is again (within the experimental error) equal to the γ-energy 637 KeV. The 175 KeV γ-line is therefore emitted between the levels at 637 and 465 KeV.

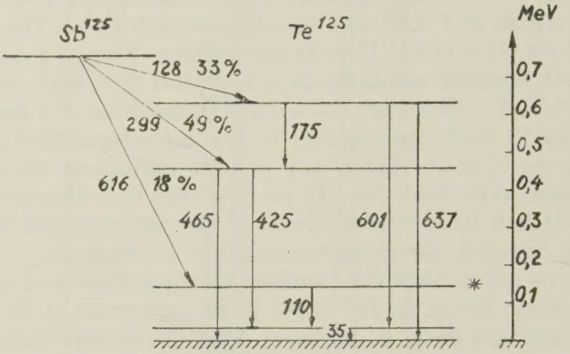

Fig. 6. Disintegration scheme for Sb^{125} and Te^{125*}.

Obviously at least one of the γ- or β-transitions must lead to the metastable level in Te. Since no γ-rays end there, it must be a β-component. If the hardest β-component ($E_\beta = 616$ KeV) goes to this level the next β-component (299 KeV) will have to end on a level with the excitation energy $35 + 110 + (616–299) = 462$ KeV. This is in very good agreement with the observed γ-ray energy of 465 KeV, emitted from this level to the ground state. The third β-component of 128 KeV will then end on the next excited level. The difference between the last two β-components is $299–128 = 171$ KeV, which again fits very well to the observed γ-line of 175 KeV.

γγ- and βγ-coincidence measurements (with and without absorbers) have been carried out. In this case it is rather difficult to compare quantitatively the expected and experimentally obtained data of the coincidence effects due to the many uncertain estimates, which have to be done. It could be concluded, however, that the results obtained from the coincidence measurements were at least consistent with the above term scheme. The main support for the scheme comes from the energetic relations discussed above.

From the intensities of the three β-components their corresponding ft values can be computed (according to KONOPINSKI's[1] notation).

The results are:

$$ft_{128} = 8.1 \times 10^6$$
$$ft_{299} = 7.6 \times 10^7$$
$$ft_{616} = 2.4 \times 10^9$$

Considering the Z-value of the nucleus in question a reasonable assignment of the degree of forbidness of the β-transitions is

128 keV β-transition	first forbidden
299 keV β-transition	second forbidden
616 keV β-transition	third forbidden

Using the selection rules for β- and γ-transitions some ideas can be obtained regarding the spins and parities of the different levels. Thus the highest excited level in Te^{125} probably has the opposite parity as the groundstate of Sb^{125} and the level next below in Te. The spin difference between this latter level and the Sb^{125} ground state (according[2] to the shell model probably $\frac{3}{2}$ or $\frac{5}{2}$) is either 2 or 3. The metastable level in Te must have the same parity as the highest excited level and a very high spin, being four units greater than the 35 KeV level etc. Since both β- and γ-selection rules always leave a number of different possibilities open, it does not seem worth while in this complicated disintegration scheme to work out further all theoretically possible spin alternatives, which are rather numerous.

SUMMARY

1. A chemical procedure is described to separate Sb^{125} and Te^{125*} with high spec. activities from a large amount of neutron irradiated tin.

2. The γ-radiation has been analyzed in a magnetic spectrograph. The following γ-lines exist: 35 KeV, 110 KeV, 175 KeV, 425 KeV, 465 KeV, 601 KeV and 637 KeV.

3. The β-spectrum has been investigated. This contains internal conversion lines, corresponding to those found in the γ-spectrum, together with one additional electron line, interpreted as an Auger line. The continuous spectrum has been resolved into three different β-components, namely $E_{\beta_1} = 128$ KeV, $E_{\beta_2} = 299$ KeV, $E_{\beta_3} = 616$ KeV with relative intensities 33 %, 49 % and 18 %.

4. The internal conversion spectrum emitted from Te^{125*} has been investigated. The K : (L + M) ratio of the 110 KeV transition is 1.10. Besides these lines two other lines have been found. One of them is a conversion line from the L shell, the corresponding γ-energy being 35 KeV, and one is an Auger electron line, showing a transition K-L-L^1.

5. A term scheme is proposed, which takes care of the experimental material obtained. This scheme is discussed. Thus it seems probable that the three β-components are (with increasing energy) 1st forbidden, 2nd forbidden and 3rd forbidden.

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[1] E. KONOPINSKI, Rev. Mod. Physics *15*, 209, 1943.

[2] E. FEENBERG, Phys. Rev. *75*, 320, 1949.

Tryckt den 8 november 1949

Uppsala 1949. Almqvist & Wiksells Boktryckeri AB